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Two-Stage Biofilter for Effective NH_3 Removal from Waste Gases Containing High Concentrations of H_2S

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ABSTRACT

A high H_2S concentration inhibits nitrification when H_2S and NH_3 are simultaneously treated in a single biofilter. To improve NH_3 removal from waste gases containing concentrated H_2S, a two-stage biofilter was designed to solve the problem. In this study, the first biofilter, inoculated with *Thiobacillus thioparus*, was intended mainly to remove H_2S and to reduce the effect of H_2S concentration on nitrification in the second biofilter, and the second biofilter, inoculated with *Nitrosomonas europaea*, was to remove NH_3. Extensive studies, which took into account the characteristics of gas removal, the engineering properties of the two biofilters, and biological parameters, were conducted in a 210-day operation. The results showed that an average 98% removal efficiency for H_2S and a 100% removal efficiency for NH_3 (empty bed retention time = 23–180 sec) were achieved after 70 days. The maximum degradation rate for NH_3 was measured as 2.35 g N day^{-1} kg of dry granular activated $carbon^{-1}$. Inhibition of nitrification was not found in the biofilter. This two-stage biofilter also exhibited good adaptability to shock loading and shutdown periods. Analysis of metabolic product and observation of the bacterial community revealed no obvious acidification or alkalinity phenomena. In addition, a lower moisture content (~40%) for microbial survival and low pressure drop (average 24.39 mm H_2O m^{-1}) for system operation demonstrated that the two-stage biofilter was energy saving and economic. Thus, the two-stage biofilter is a feasible system to enhance NH_3 removal in the concentrated coexistence of H_2S.

IMPLICATIONS

A two-stage biofilter was applied for the first time to sequentially treat concentrated H_2S and diluted NH_3 mixtures. Essential operating characteristics, including system performance, engineering properties, and biological parameters, were presented to illustrate the feasibility of the system. High NH_3 removal efficiency compared with a conventional single biofilter was achieved for a long operating period.

INTRODUCTION

Large amounts of NH_3 and H_2S are often simultaneously expelled from various industrial processes and sources, such as petrochemical refining, food preparation, metallurgy, wastewater treatment, treatment of fuels, livestock farming, and hog manuring.[1–4] Mixed gases containing concentrated H_2S (>200 ppmv) and diluted NH_3 (<80 ppmv) exist in some specific emission sources like wastewater lift stations, sewage treatment plants, and night soil treatment plants.[5–7] These emissions, in addition to their own toxicity, constitute a source of olfactory nuisance.

To control the emissions of high-concentration gaseous pollutants (>5000 ppmv), incineration and dielectric barrier discharges (DBDs) are generally given priority.[8] Although these technologies are effective at controlling odor-causing substances, they produce high treatment costs and secondary waste stream problems. Traditional waste gas treatments, such as activated carbon adsorption, wet scrubbing, and masking, have been used to remove relatively low-concentration gaseous pollutants from waste gases. These methods, however, often transfer odor-causing materials from gas phase to scrubbing liquids or solid adsorbents, and their derivatives resulted from wastewater and solid waste questions.[9,10] Generally, sludge bioreactor systems for removing odor-causing pollutants are acceptable, but the relatively long residence times required to achieve a high removal efficiency (e.g., >95%) and the relatively large reactor footprints required because the systems do not use a specific microbial inoculant often limit their application.[11] As regulatory measures move toward more stringent control of gaseous pollutants, the development of more cost-effective and efficient air pollution control technology grows ever more important. Recently, biofiltration has been regarded as the best available control technology for treating diluted pollutants and odorous compounds.[12] The technique contains higher microbial content, environmental endurance, and operational stability at a lower treatment cost.[13] However, high H_2S concentrations (120 ppmv) significantly suppress the NH_3 removal from waste gases in a coimmobilized cell biofilter.[14] Similar results have also been found by Kim et al.[15] and Lee et al.[16] This dramatic reduction in NH_3 removal is because of the simultaneous introduction of high H_2S concentrations, which result in

Table 1. Composition of basal media for cultivation of microorganisms and continuous experiments.

Medium	Ammonia Medium		Thiosulfate Medium		Inflow Medium	
Composition (g L^{-1})	$(NH_4)_2SO_4$	3.3	NH_4Cl	0.4	NH_4Cl	0.01
	$MgSO_4$	0.25	$MgCl_2{\cdot}6H_2O$	0.2	$MgCl_2{\cdot}6H_2O$	0.2
	NaH_2PO_4	0.78	NaH_2PO_4	1.2	NaH_2PO_4	0.78
	Na_2HPO_4	0.89	Na_2HPO_4	1.2	Na_2HPO_4	0.92
	$CaCl_2$[a]	0.74	$Na_2S_2O_3{\cdot}5H_2O$	8.0	$CaCl_2$[a]	0.74
	$FeSO_4{\cdot}7H_2O$[a]	2.5	$FeSO_4{\cdot}7H_2O$	0.01	$FeSO_4{\cdot}7H_2O$	0.01
	$CuSO_4$[a]	0.08			$CuSO_4$[a]	0.08
pH	7.5		7.0		7.5	
Temperature (ºC)	30		30		30	

Notes: [a]mg L^{-1}.

a large accumulation of acidic metabolites and creates substantial operational problems for traditional biofilters. Declining pH in the biofilter substantially inhibits and hinders the nitrification.[17] Thus, to overcome these defects, the choice of an appropriate packing material, the screening of acid-tolerant nitrifying bacteria, and an improvement in the biofilter type should be feasible strategies. Granular activated carbon has been considered as a packing material for biofilter systems,[15,18,19] because it provides a more uniform surface area, good resistance to crushing, and good alkalinity for neutralizing acid gases.[20] Combined with a microbial oxidation process, pollutant-laden activated carbon can be autoregenerated by bacterial cells residing on or in it, which is another advantage.[19]

Nitrosomonas europaea is effective in removing NH_3 and *Thiobacillus thioparus* in eliminating H_2S.[21,22] Therefore, these two species were chosen to inoculate the biofilter. In this study, a two-stage biofilter packed with activated carbon was designed to clarify concentrated H_2S and diluted NH_3 gas mixtures for 210 days. The two-stage biofilter containing two single biofilters in series was used to enhance the efficiency of the nitrification by removing H_2S in the first biofilter. The purpose of the first biofilter, inoculated with *T. thioparus*, was mainly to remove H_2S. The second biofilter, inoculated with *N. europaea*, was to remove NH_3. The goal was to effectively remove NH_3 from waste gases with high H_2S concentrations.

EXPERIMENTAL WORK

Organism Cultivation and Medium Preparation

The original pure-culture strains of autotrophic sulfur oxidizer (*T. thioparus*) and autotrophic ammonia oxidizer (*N. europaea*) were isolated from swine wastewater.[21,22] The stock cultures of *T. thioparus* were grown in the thiosulfate medium at 30 °C with 120 strokes min^{-1}. The stock cultures of *N. europaea* were grown in the ammonia medium in the dark at 30 °C with 120 strokes min^{-1}. For all of the continuous experiments, the inflow medium was provided from the nutrient tank. The compositions of these basal media are listed in Table 1.[14] The final pH of the medium was adjusted using 2 N sodium hydroxide or hydrochloric acid.

Immobilization Procedure

Commercially available granular activated carbon (Taiwan Activated Carbon Industries Company) obtained from coconut shell and with a particle size of 4.5 mm was used as the support material for the biomass in this study. The main characteristics of the support material (bulk density, specific surface area, and pH value; provided by the vendor) were 0.48 g cm^{-3}, 1250 m^2 g^{-1}, and 9, respectively. *N. europaea* grown in 100 mL of ammonia medium and *T. thioparus* grown in 100 mL of thiosulfate medium for 5 days were harvested by centrifugation (7500 × *g* for 10 min) and then washed thrice with sterile phosphate buffer. The cell precipitate of *N. europaea* and *T. thioparus* was separately drawn and put into a 10-L polyvinyl chloride tank containing 7 L of ammonia and thiosulfate medium, respectively. Before initiating the biofilter experiments, 2.7 kg of the granular activated carbon (GAC) were separately mixed with the above solution for microbial attachment. During the immobilization period, the ammonia and thiosulfate liquid media were replaced every 2 weeks until the cell numbers 10^9–10^{10} CFU (colony forming units)/g dry GAC^{-1} were achieved for both *N. europaea* and *T. thioparus*. After 2 months, the cell-laden GAC in the tank was separately packed into two biofilter columns. All of the materials and implements were maintained in aseptic conditions during the above experimental period.

Apparatus and NH_3/H_2S Sequential Removal in the Continuous Operation

To investigate the capacity of GAC to adsorb NH_3 and H_2S, respectively, three glass columns (12 cm ϕ × 5 cm working height) connected in series were packed with GAC without microbes to perform the bed depth service time experiment.[23,24] NH_3 or H_2S (60 ppmv) was continuously supplied to these glass columns at 500 L hr^{-1}. The desired concentration of NH_3 or H_2S at the breakthrough was defined as 12 ppmv. The adsorption process can be described in the following equation[24]:

$$t = \left(\frac{N_0}{C_0 V}\right) X - \frac{1}{KC_0} \ln\left(\frac{C_0}{C_e} - 1\right) \qquad (1)$$

where C_O and C_e are the inlet concentration and the desired concentration (ppmv) of gas at breakthrough, respectively; V, t, and X are hydraulic loading (cm hr^{-1}), service time (hr), and packing bead depth (cm), respectively; and N_0 and K are adsorptive loading (ppmv) and adsorption rate constant (hr^{-1} $ppmv^{-1}$).

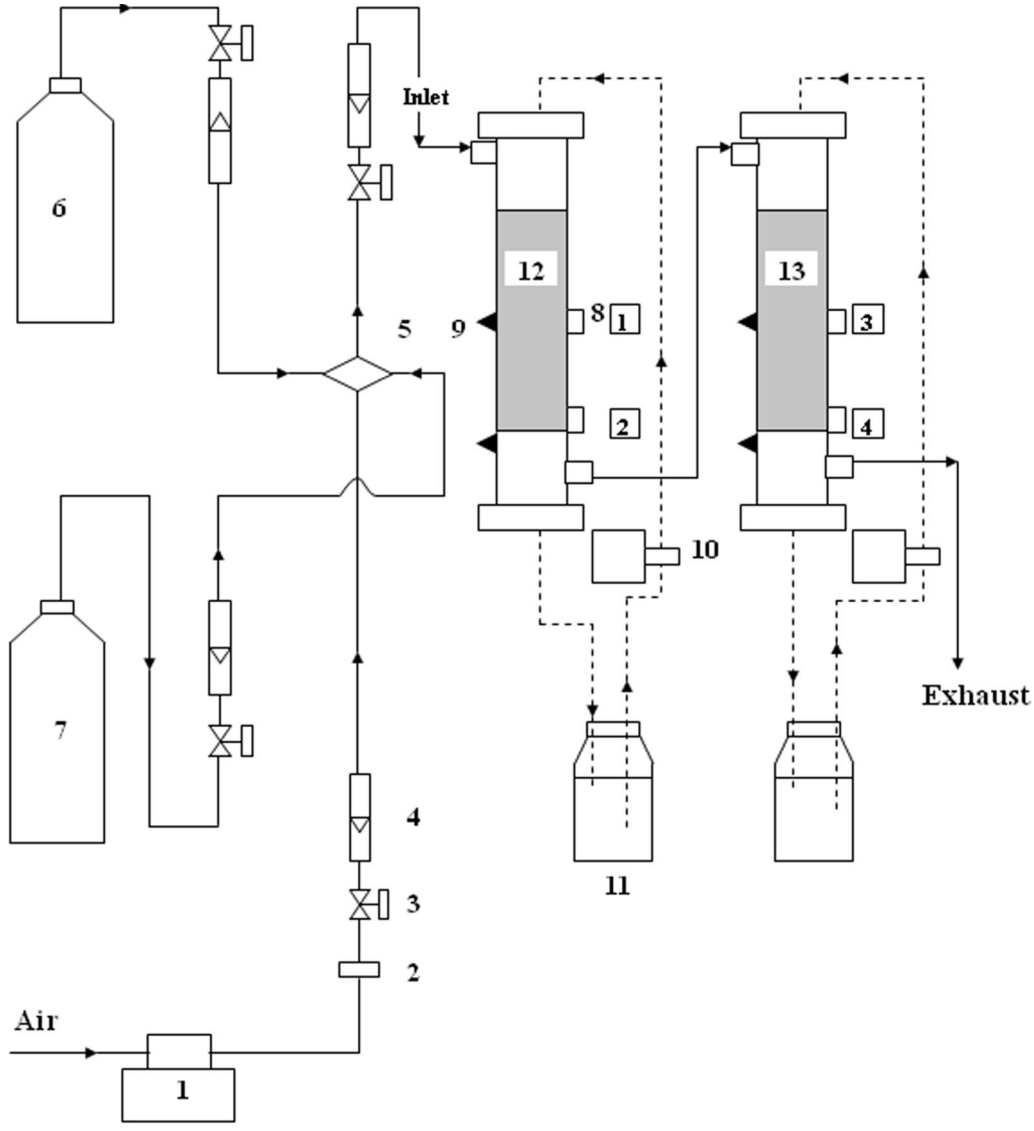

Figure 1. Schematic of the two-stage biofilter. 1 = air compressor; 2 = air filter; 3 = regulator; 4 = flow meter; 5 = four-way connector; 6 = H_2S gas cylinder; 7 = NH_3 gas cylinder; 8 = medium sampling port; 9 = gas sampling port; 10 = peristaltic pump; 11 = nutrient tank; 12 = biofilter inoculated with *T. thioparus*; 13 = biofilter inoculated with *N. europaea*.

The setup and design of the two-stage biofilter is shown in Figure 1 and can be illustrated as follows. Two glass columns (each column: 12 cm $\phi \times 40$ cm of working height) connected in series were separately packed with immobilized-cell GAC, and a perforated sieve plate of polyvinyl chloride material was fitted at the bottom of these columns to allow the circulating liquid to flow out. To enhance NH_3 removal in the coexistence of concentrated H_2S, the first-stage biofilter was packed with the immobilized-cell GAC of *T. thioparus*, and the second stage biofilter was packed with the immobilized-cell GAC of *N. europaea*. The packed volume and GAC dry weight in each biofilter were 4.52 L and 2.17 kg, respectively. Sampling ports spaced 20 cm apart were installed along the biofilter to measure gas concentrations and the composition of packing material. The first stage biofilter had two sampling ports, named location 1 (30 cm from the inlet) and location 2 (50 cm from the inlet), respectively. Similarly, the second stage biofilter had two sampling ports, named location 3 (90 cm from the inlet) and location 4 (110 cm from the inlet), respectively. The flow meters and valves were used for monitoring and controlling the gas flow through the biofilter. The biofilter was wrapped in a heating blanket to maintain its temperature at 30 ± 2 °C. The pure NH_3 and H_2S gases (purity: 99.999%), supplied from separate gas cylinders (San Fu Chemical Co), were first diluted with compressed air, which had passed through an air filter (pore size 0.2 μm, LIDA 3000-06). The diluted gasses then flowed downward through the top of the biofilter's first stage. The exhaust of the first-stage biofilter was directed through the biofilter's second stage the same way. The inflow media (Table 1) separately stored in the nutrient tanks of the first- and second-stage biofilters were recirculated by peristaltic pumps at 5 L

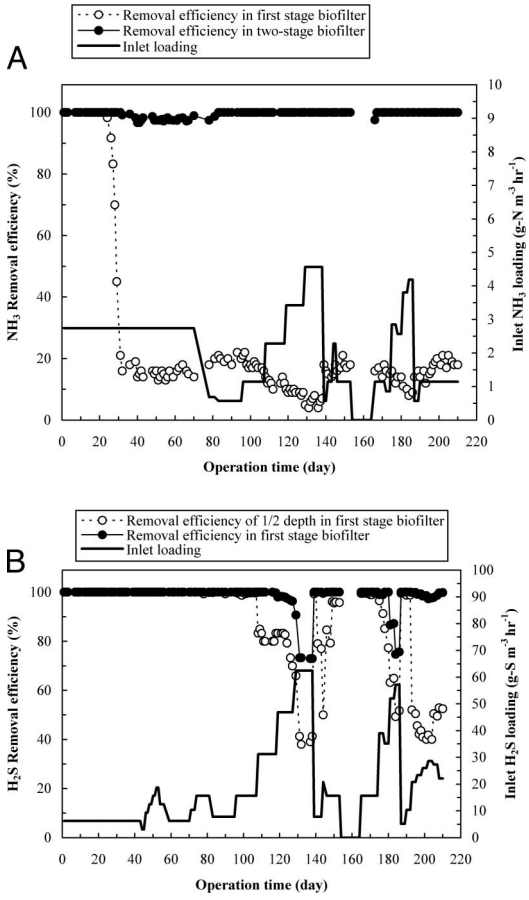

Figure 2. (A) Removal efficiency and inlet loading profiles of NH_3 in the two-stage biofilter under NH_3 and H_2S mixed-waste gases during the 210-day operation at 30 ± 2 °C. (B) Removal efficiency and inlet loading profiles of H_2S in the two-stage biofilter under NH_3 and H_2S mixed-waste gases during 210-day operation at 30 ± 2 °C.

min^{-1} for 2 min six times a day to maintain the moisture of the two-stage biofilter and supply nutrient to the attached cells. The peristaltic pump was connected to a spray nozzle located on the top of the filter to uniformly spray the medium on the filter bed surface. Every 3 months, the inflow medium was replaced with fresh medium unless stated otherwise.

To examine the operating performance of the two-stage biofilter in response to different ratios of an NH_3/H_2S gas mixture, the operation periods were designed for two phases. In the first phase (0–70 days), various concentrations of NH_3 (30–120 ppmv) and H_2S (30–180 ppmv) were simultaneously introduced to the biofilter system at 180-1440 L hr^{-1} (empty bed retention time [EBRT]: 23–180 sec). In the second phase (71–210 days), 60 ppmv of NH_3 and high H_2S concentrations (200–300 ppmv) were simultaneously introduced to the biofilter system at 180-1440 L hr^{-1}. To evaluate the effect of an intermittent operation on the removal efficiency of the system, common air without NH_3 or H_2S pollutant was supplied on days 154–163 (see Figure 2, A and B).

Kinetic Analysis

The removal rate of NH_3 in the second-stage biofilter was evaluated using a Michaelis–Menten equation.[14] In this case, the distribution of gas flow in the biofilter was taken as the plug flow, and the process could be described in the following equation:

$$\frac{1}{R} = \frac{K_s}{V_m} \times \frac{1}{C_{ln}} + \frac{1}{V_m} \quad (2)$$

where R (g-N day^{-1} kg GAC^{-1}) = (Q × C_o × R%)/W, the apparent removal rate calculated by gas flow rate (Q), inlet concentration (C_o), gas removal efficiency (R%), and GAC weight (W); C_{ln} (ppmv) = (C_o − C_e)/ln (C_o/C_e), logarithmic mean concentration of NH_3 at the inlet and outlet of second-stage biofilter; V_m (g-N day^{-1} kg GAC^{-1}) is the maximum apparent removal rate, and K_s (ppmv) is the apparent half-saturation constant. From the linear relationship between $1/C_{ln}$ and 1/R, V_m and K_s were calculated from the slope and intercept. In this experiment, the flow rates were controlled in the range of 180–360 L hr^{-1} (EBRT: 90–180 sec) to minimize the external mass-transfer limitation. The waste gases containing 300 ppmv H_2S and NH_3 concentrations ranging from 30–120 ppmv were fed into the biofilter system.

Analytical Methods

Inlet and outlet H_2S gas concentrations in the biofilter were measured either continuously by a single point monitor[25] (MDA Scientific) ranging from 50–1500 ppb or periodically by gas detector tubes (GASTEC) ranging from 12.5 to 500 ppmv or 1 to 40 ppmv. Inlet and outlet NH_3 gas concentrations were measured either continuously by a single point monitor[25] (MDA Scientific) in the range of 0.1–10 ppmv or periodically by gas detector tubes (GASTEC) in the range of 2.5–200 ppmv. The ranges of error for the single point monitor were 2.5% and 5% for H_2S and NH_3, respectively. The ranges of error for the gas detector tubes were 5% and 10% for H_2S and NH_3, respectively. In all of the continuous experiments, the variation in H_2S and NH_3 concentrations at steady state was within ±5%. Therefore, the 12 values obtained at steady states were averaged as the H_2S or NH_3 outlet concentration. Samples were taken 48 times per day for periodic measurement with the gas detector tubes.

To determine the pressure drop across the two-stage biofilter under different flow rates, water-filled manometers were applied during the continuous operation period, and the unit was expressed as mm-H_2O m-filter $height^{-1}$. To determine the moisture content in the GAC, ~2 g of GAC were withdrawn, weighed, and dried over a 24-hr period at 103 ± 0.5 °C. To avoid a superposition by a temporal effect, the measurements of pressure drop and moisture versus flow rate were conducted after the flow rate had been stable for 5 days and liquid recirculation had occurred after 2 hr. Additionally, randomly chosen flow rate measurements were conducted. To measure the pH of GAC, 0.5 g of GAC were withdrawn from the biofilter through the appropriate sampling ports and mixed with 5 mL of distilled water. The sample was vortexed for 3 min, and the pH was then determined using a pH meter.[26] For microbial analyses, 0.5 g of GAC were separately taken from sampling ports at the different depths and mixed with 5 mL of sterile phosphate buffer.

These samples were vortexed for 3 min, and the resulting liquid with different dilution ratios was spread on the solid ammonia and thiosulfate medium, respectively. After incubation for 5 days at 30 °C, the numbers of colonies on the plates were counted. To analyze these metabolic products, 1 g of GAC was withdrawn from the biofilter and mixed with 10 mL of distilled water. The sample was vortexed for 3 min, and the GAC was withdrawn to analyze the elemental sulfur (S). The residual suspension solutions were analyzed for other chemical compositions. Sulfate ($SO^{=}$), nitrate (NO_3^-), and nitrite (NO_2^-) concentrations in the solution were measured by ion chromatography (Dionex 4500i). Ammonium (NH_4^+) and sulfide ($S^{=}$) were determined using an ion-specific electrode. Sulfite ($SO_3^{=}$) was determined by titration using a standard potassium iodide-iodate titrant and a starch indicator.[22] Elemental sulfur was determined by reacting it with cyanide to produce thiocyanate, which was then quantified as $Fe(SCN)_6^{-3}$.[22] Organic nitrogen was determined by the Kjeldahl method.[27]

RESULTS AND DISCUSSION

NH_3/H_2S Sequential Removal in the Continuous Operation

The NH_3 and H_2S removal efficiency profiles in the two-stage biofilter during 210 days of operation are indicated in Figure 2, A and B. According to the calculations from eq 1, the adsorptive loading and adsorption rate constant of GAC for NH_3 were 281 ppmv and 1.7 hr^{-1} $ppmv^{-1}$, respectively. These values for H_2S were 5057 ppmv and 70.3 hr^{-1} $ppmv^{-1}$, respectively. The theoretical saturation times were ~12 and 47 days for single NH_3 and H_2S adsorption, respectively, by estimating real operating conditions (flow rates and concentrations). The adsorption capacity of H_2S is greater than NH_3 by pure GAC. Saturation adsorption time is shortened by NH_3 and H_2S coexistence. However, the removal efficiency profiles indicated that 100% removal efficiencies of NH_3 and H_2S in 32 and 78 days were achieved, respectively. These results suggested that NH_3 and H_2S were removed, not only through adsorption by GAC, but also through biooxidation by microorganisms.

To apply the two-stage biofilter to various emission sources, the operation periods were designed for two phases. In the first phase (0–70 days), various concentrations of NH_3 (30–120 ppmv) and H_2S (30–180 ppmv) were introduced to system. In this operation period, H_2S was completely removed in the first stage. A removal efficiency for NH_3 >97.5% was achieved in the two-stage biofilter, and 21% NH_3 was removed at most in the first stage after saturation of GAC adsorption. Apparently, GAC adsorption was responsible for NH_3 removal in the first stage, which may have resulted from the fact that *N. europaea* was not inoculated into the column. In the second phase (71–210 days), 60 ppmv of NH_3 and 200–300 ppmv of H_2S were simultaneously fed to the biofilter system at 180-1440 L hr^{-1}. Figure 2, A and B, indicates that variations of inlet H_2S loading (from 5.2 to 62.5 g-S m^{-3} hr^{-1}) were higher than variations of inlet NH_3 loading (from 0.57 to 4.56 g-N m^{-3} hr^{-1}). When the H_2S inlet loading gradually increased from 7.8 g-S m^{-3} hr^{-1} (on day 95) to 62.5 g-S m^{-3} hr^{-1} (on day 132), the removal efficiencies of H_2S plunged from 100% to 73% because of lack of gas retention time. Once the inlet H_2S loading came back to 7.8 g-S m^{-3} hr^{-1}, complete H_2S removal was immediately achieved because of sufficient gas retention time and high microbial activity. The second shock loading was performed during days 170 to 185, and the system exhibited a similar adaptability. In the second phase, an average of 98.2% H_2S could be removed. According to a previous study by Chung et al.,[14] coexistence of high H_2S significantly suppresses NH_3 removal from waste gases. However, even if 200–300 ppmv of H_2S was introduced to the two-stage biofilter, NH_3 removal efficiency of ~100% was achieved, because almost all of the H_2S had been removed in the biofilter's first stage except during the shutdown period. Apparently, the modified biofilter has improved NH_3 removal efficiency in the coexistence of concentrated H_2S. Previous reports have shown that the removal efficiencies of NH_3 in simultaneously purifying concentrated H_2S and diluted NH_3 in a wood chip biofilter, a GAC biofilter, a ceramic biofilter, a coimmobilized cells biofilter, and a fluidized bed biofilter were 50%, 54%, 90%, 95%, and 98%, respectively.[7,14–16,28] In comparison with these results, the two-stage biofilter indeed improved on NH_3 removal from waste gases containing high H_2S concentrations under similar conditions.

Effect of Intermittent Operation

Emissions of waste gases are not always continuous in the manufacturing and production processes of various industries. Hence, the influence of intermittent operation on the two-stage biofilter is examined. During the 210-day operation, a 10-day shutdown period (day 2, 154–163) was carried out. The common air and inflow medium were continuously provided, but no NH_3 and H_2S gases were fed. When the mixtures containing NH_3 and H_2S were introduced into the system again after 10 days, the removal efficiency of NH_3 immediately reached 97.6% on the first day and 100% on the third day (Figure 2A). Additionally, the removal efficiency of H_2S reached 100% in the biofilter's first stage on the first day and 99.5% H_2S removal at the half depth of the first stage on the first day (Figure 2B). Compared with the removal efficiency of H_2S at the half depth of the first-stage biofilter before or after intermittent operation, the removal efficiency after intermittent operation rose significantly ($P < 0.05$) in the initial 6 days (Figure 2B). The results seemed counterintuitive. However, when we determined the bacteria numbers before or after intermittent operation (see Figure 6), the difference was insignificant. Because *N. europaea* and *T. thioparus* are autotrophic bacteria, their counts should fall significantly under operating conditions lacking NH_3 or H_2S. Hence, the phenomenon must have been because of the fact that NH_3 and H_2S absorbed on GAC could be continuously supplied to *N. europaea* and *T. thioparus* during the shutdown periods. This could be proven by the continuous increase in the metabolic product (NO_2^- or $SO_4^{=}$) of NH_3 and H_2S produced by *N. europaea* and *T. thioparus*. In the biofilter's first stage, the sulfate concentrations in the effluent on days 154, 158, and 163 were 3.2 g L^{-1}, 3.8 g L^{-1}, and 4.3 g L^{-1}, respectively. In the second stage, the nitrite concentrations in the effluent on days 154, 158, and 163 were 0.21 g L^{-1}, 0.26 g L^{-1}, and

Table 2. Sulfur mass balances at the half depth of the first-stage biofilter during the 210-day operating period.

Species	H_2S Removed (g S kg dry GAC^{-1})	$SO_4^=$ (g S kg dry GAC^{-1})	S^o (g S kg dry GAC^{-1})	$SO_3^=$ (g S kg dry GAC^{-1})	$S^=$ (g S kg dry GAC^{-1})
Amount	584.2	172.1	382.4	1.6	5.6
Ratio[a]	–	30.6%	68.1%	0.3%	1.0%

Notes: [a]Conversion ratio of H_2S removed to varied products.

0.31 g L^{-1}, respectively. Hence, GAC unsaturated by NH_3 or H_2S after intermittent operation provided more adsorption locations for pollutants than before intermittent operation. Thus, a high removal efficiency by the system was still achieved even after the 10-day shutdown period. These operating characteristics showed that the two-stage biofilter could cope with shock loading and intermittent operation and still provide effective NH_3 removal in the coexistence of high H_2S concentrations.

Metabolic Products Analysis

To understand the metabolic products of NH_3 and H_2S gases by *N. eurpoaea* and *T. thioparus*, mass balances of sulfur and nitrogen in the two-stage biofilter were conducted after 210 days of operation, and they are listed in Tables 2 and 3. GAC (1 g) was withdrawn from the middle of the first- and second-stage biofilter for all of the product analyses. As Table 2 indicates, the total amount of H_2S consumption was 584 g S kg dry GAC^{-1}, and the $SO_4^=$, S^o, $SO_3^=$, and $S^=$ accumulations were 172.1, 382.4, 1.6, and 5.6 g S kg dry GAC^{-1}, respectively. Therefore, the conversion ratios of H_2S to $SO_4^=$, S^o, $SO_3^=$, and $S^=$ were 30.6%, 68.1%, 0.3%, and 1%, respectively. The determined products accounted for 96.1% (561.7/584.2) of the total H_2S conversion. The remaining 3.9% of the conversion product may exist in the recycling solution or be transformed to organic sulfur. Because the major metabolic products were neutral S^o (68%), the pH at the half depth of the first-stage biofilter stayed within the neutral range, except under specific conditions (see Figure 6). Similar results also were found in our previous research.[19] Table 3 shows that the distribution of the total nitrogen at the half depth of the second-stage biofilter was divided into four species at least. The conversion ratios of NH_3 to NO_3^-, NO_2^-, NH_4^+, and organic N were 10.2%, 82.4%, 6.1%, and 1.3%, respectively. These products accounted for 92.5% (378.4/409.1) of the total NH_3 conversion. The remaining parts should exist in the recycling solution. These data indicated that the NH_3 metabolism was responsible for the formation of NO_2^-.[14] A smaller amount of NH_4^+ (or NH_3) was observed, because it was adsorbed on GAC bead or absorbed in the surrounding H_2O of GAC. This could provide partial alkalinity to neutralize acidity derived from some acidic products and maintain the pH stability of the system. However, because nitrite is the main product of ammonia metabolites, this means that nitrification is not complete. Nitrite accumulation might pose additional problems, because it may hamper biological treatment of spent nutrient solution in a large-scale application of the process. Thus, inoculating nitrite-oxidizing bacteria may be a countermeasure to take in the future.

Effect of H_2S Concentration on NH_3 Removal

To investigate the effect of H_2S concentration on NH_3 removal from waste gases containing NH_3 and H_2S, different H_2S concentrations (30–300 ppmv) were introduced at 720 L hr^{-1} (EBRT: 45 sec) accompanied by 60 ppmv NH_3. The NH_3 removal ratios and NH_3 concentrations versus position for the different inlet H_2S concentrations in the two-stage biofilter are shown in Figure 3. A <3% NH_3 removal ratio was achieved at locations 1 and 2 in the first stage. Conversely, relatively high NH_3 removal ratios (41–52%) were observed at locations 3 and 4 in the second stage. Because this stage was inoculated with ammonia-oxidizing bacteria, the results were reasonable. Additionally, the NH_3 removal ratio was not influenced by H_2S concentration in statistical analysis. The average removal efficiency of NH_3 in the two-stage biofilter was 99.3%. In the previous study, 120 ppmv of H_2S would significantly affect NH_3 removal when these gases coexisted in the biofilter.[14] However, in the two-stage biofilter, even 300 ppmv of H_2S did not affect NH_3 removal. Because the first stage biofilter inoculated with *T. thioparus* was responsible for H_2S degradation, only 2.5% of the H_2S introduced to stage 1 (i.e., max 7.5 ppmv) entered the second-stage biofilter, where interference with nitrification was not to be expected.

To clearly illustrate the fact, a kinetic analysis was conducted. The apparent kinetic parameters of the maximum removal rate and half-saturation constant to degrade NH_3 (30–120 ppmv) under 300 ppmv H_2S coexistence were evaluated by the Lineweaver-Burk method, and the results are shown in Figure 4. The regression equation

Table 3. Nitrogen mass balances at the half depth of the second stage biofilter during the 210-day operating period.

Species	NH_3 Removed (g N kg dry GAC^{-1})	NO_3 (g N kg dry GAC^{-1})	NO_2 (g N kg dry GAC^{-1})	NH_4^+ (g N kg dry GAC^{-1})	Organic N (g N kg dry GAC^{-1})
Amount	409.1	38.6	311.8	23.1	4.9
Ratio[a]	–	10.2%	82.4%	6.1%	1.3%

Notes: [a]Conversion ratio of NH_3 removed to varied products.

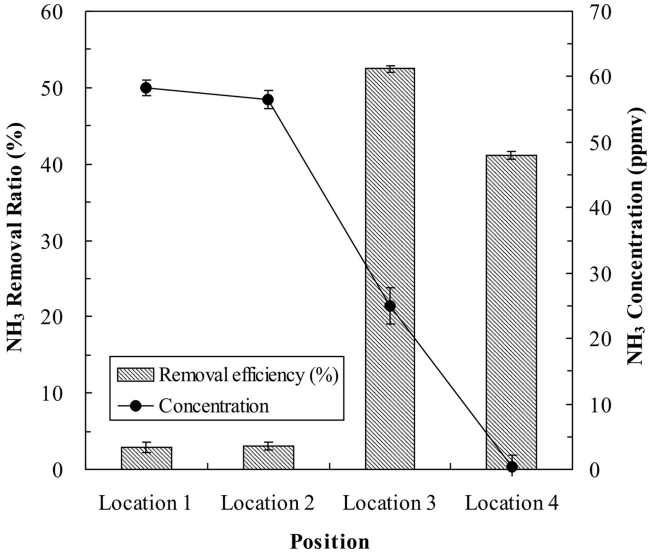

Figure 3. The NH_3 removal ratios and NH_3 concentrations vs. position for different inlet H_2S concentrations in the two-stage biofilter. The H_2S concentrations ranged from 30 to 300 ppmv, and 60 ppmv of NH_3 were simultaneously introduced at 720 L hr^{-1}. The error bars indicate the standard deviation.

was expressed as $y = 41.248x + 0.4242$. The maximum removal rate and the half-saturation constant for NH_3 were calculated to be $V_m = 2.35$ g N day^{-1} kg dry GAC^{-1} and $K_s = 17.5$ ppmv from the slope and intercept of the regression equation. The saturation constant in this study was lower than the values of 49.9 and 34.9 ppmv by *N. europaea* reported by Bedard et al.[29] and Keener et al.[30] and much lower than the value of 83.1 ppmv by *Arthrobacter oxydans*.[23] If we supposed a physical meaning of K_s analogous to that in enzyme kinetics, a decrease in K_s suggests an enhancement in biomass affinity for NH_3 removal. Therefore, *N. europaea* in the two-stage biofilter apparently exhibited higher nitrification activity than other nitrification biofilters. A control test without simultaneous feeding of H_2S gas was also examined. The differences between the kinetic parameters (K_s and V_m) of the control test and the H_2S degradation test were not significant ($P > 0.05$). To more clearly illustrate the kinetics in terms of a different kinetics order, we also analyze the kinetics change with inlet NH_3 concentration. When inlet concentrations were >100 ppmv, the reaction followed zero-order kinetics. When inlet concentrations were <50 ppmv, the reaction followed first-order kinetics. When inlet concentrations fell into the intermediate range, the reaction followed intermediate kinetics. These results suggest that using the two-stage biofilter will improve NH_3 removal from waste gases containing high H_2S concentrations.

Effect of Empty Bed Retention Time on NH_3 and H_2S Removal

The effect of empty bed retention time in the two-stage biofilter on H_2S and NH_3 removal was studied by simultaneously introducing 60 ppmv of NH_3 and 300 ppmv of H_2S to the system at various gas flow rates. The removal efficiencies of H_2S and NH_3 were separately evaluated in the biofilter's first stage and second stage, and the results are shown in Figure 5. Removal efficiency decreased progressively with decreasing retention time (increasing gas flow rate).

Complete removal efficiencies of NH_3 and H_2S gases were achieved when the retention times were >36 sec and 45 sec, respectively. When the retention times of NH_3 and H_2S were >22 and 30 sec, a >97% removal efficiency was achieved. The results indicate that H_2S removal seemed more sensitive to empty bed retention time than NH_3 removal in the biofilter system. The results may be because of lower solubility for H_2S than NH_3. Once the retention time of H_2S gas was shortened to 25 sec, the removal efficiency of H_2S plunged to 75%. Compared with previous studies, our results suggest that the two-stage biofilter possesses a removal efficiency superior to

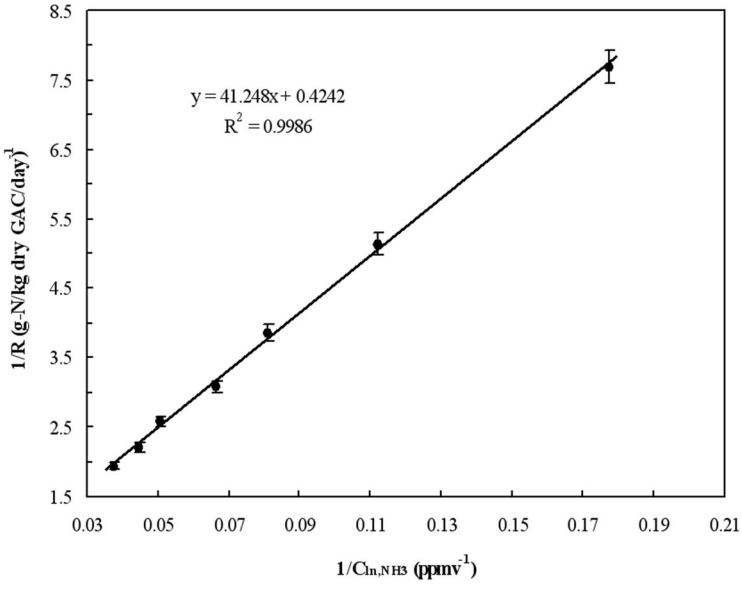

Figure 4. Relationship between 1/R and $1/C_{ln}$ of NH_3 degradation in the two-stage biofilter. The NH_3 ranged from 30 to 120 ppmv, and 300 ppmv of H_2S were simultaneously introduced to the system under flow rates of 180–360 L hr^{-1}. The error bars indicate the standard deviation.

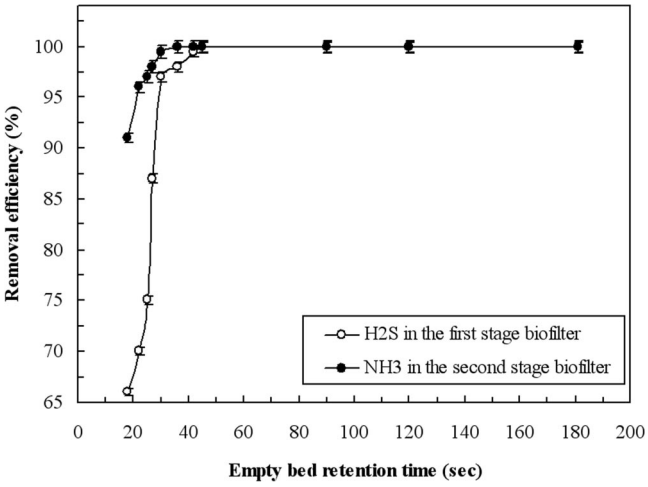

Figure 5. Effect of empty bed retention time on H_2S and NH_3 removal efficiency in the two-stage biofilter. NH_3 and H_2S were controlled at 60 ppmv and 300 ppmv at 30 ± 2 °C, respectively. The error bars indicate the standard deviation.

coimmobilized cell biofilters packed with similar autotrophic bacteria[14] and coimmobilized cell biofilters packed with heterotrophic bacteria[28] in treating waste gases containing NH_3 and H_2S under similar EBRTs.

Change in Cell Number and pH Value in the Two-Stage Biofilter

To understand the microbial activity and its distribution in the two-stage biofilter, the changes in the cell numbers were periodically examined, and the results are shown in Figure 6. During the 210-day experimental periods, the ammonia, thiosulfate solid media, and nutrient agar were used to measure the changes in the cell number of *T. thioparus*, *N. europaea*, and heterotrophic bacteria. The dominant species was *T. thioparus* in the first stage and *N. europaea* in the second stage. Other species were not observed, because their counts were below detection limits

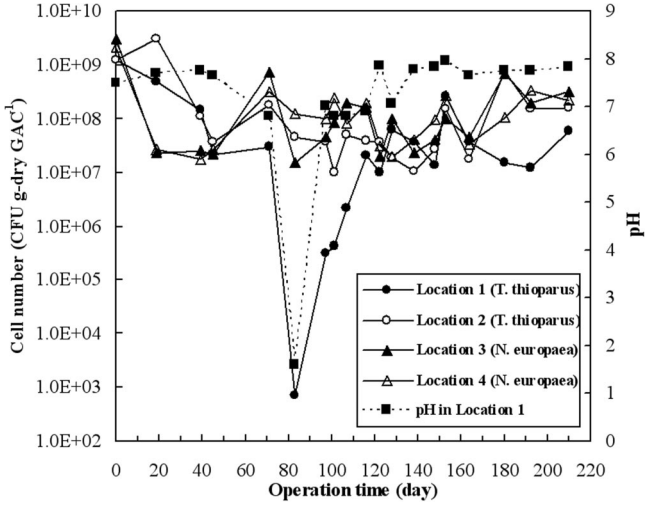

Figure 6. Change in cell number and pH value at different sampling ports in the two-stage biofilter during a 210-day operation period at 30 ± 2 °C. Locations 1 and 2 were in the first-stage biofilter, and locations 3 and 4 were in the second-stage biofilter.

(10^2 colony forming units [CFU] g dry GAC^{-1}). The experimental results indicated that the variation in the cell number of *T. thioparus* or *N. europaea* nearly ranged between 10^7 and 10^{10} CFU g dry GAC^{-1} at the different sampling ports during long-term operation. However, the cell number of *T. thioparus* in location 1 (half the height of first-stage biofilter) was reduced from 2.9×10^7 CFU g dry GAC^{-1} on day 71 to 7.1×10^2 CFU g dry GAC^{-1} on day 83 accompanying a pH drop to 1.6 (Figure 6). The dramatic reduction in cell number of *T. thioparus* during the period may be because of sudden H_2S shock loading (Figure 2B) or acidification of the first-stage biofilter.[14] Because the packing material in locations 2–4 received lower H_2S concentrations than that at location 1, no acidification was observed. On day 84, the inflow medium of the first-stage biofilter was replaced with fresh inflow medium containing a higher phosphate buffer concentration (50 mM), and the pH of GAC was restored to neutral on day 97. Apparently, the pH adjustment using phosphate buffer worked. Until experiment end, the pH of GAC at location 1 stabilized in the range of 6.8–7.9 without any acid or base addition, although shock loading occurred twice on the days 107–129 and 170–185. Similarly, the cell number of *T. thioparus* at location 1 gradually increased to 2.1×10^7 CFU g dry GAC^{-1} on day 116 and then was stable at $\sim 3.5 \times 10^7$ CFU g dry GAC^{-1}. Apparently, the reduction in cell number of the biofilter was mainly because of pH drop, not H_2S shock loading. In other words, a high H_2S concentration itself did not inhibit growth of *T. thioparus*, but the decrease in pH value was a key factor. As for *N. europaea*, its cell number showed a high stability (10^7–10^8 CFU g dry GAC^{-1}) during the 210-day operation except during the initial operation. Because physical adsorption was responsible for the initial several days in the first stage, a lower NH_3 concentration was fed into the second stage during this operating period, which reduced the cell numbers of autotrophic *N. europaea* from 3.1×10^9 to 2.3×10^7 CFU g dry GAC^{-1} at location 3 and from 2.1×10^9 to 2.7×10^7 CFU g dry GAC^{-1} at location 4. In addition, the growth of *N. europaea* was not inhibited under the coexistence of high H_2S concentrations as in our previous study,[17] and this is an additional advantage to using the two-stage biofilter.

During the 10-day intermittent operation (days 154–163), the change in cell number of the GAC in the system is also shown in Figure 6. The cell numbers of *T. thioparus* and *N. europaea* fell slightly, but a statistically insignificant difference was found between the cell number before and after intermittent operation. Because these autotrophic bacteria could use H_2S and NH_3 gases that adsorbed on GAC as their energy source during the 10-day intermittent operation, *T. thioparus* and *N. europaea* could survive. In addition, the concentrations of their metabolic products (NO_2^- and $SO_4^=$) constantly increased, and NH_4^+ constantly decreased from 0.64 g L^{-1} to 0.28 g L^{-1} in the effluent liquid, proving that *T. thioparus* and *N. europaea* were still active in the two-stage biofilter during the intermittent period.

Moisture Changes in the Two-Stage Biofilter

Microbial activity and gas mass-transfer rate in the biofilter often depend on the moisture content in or around

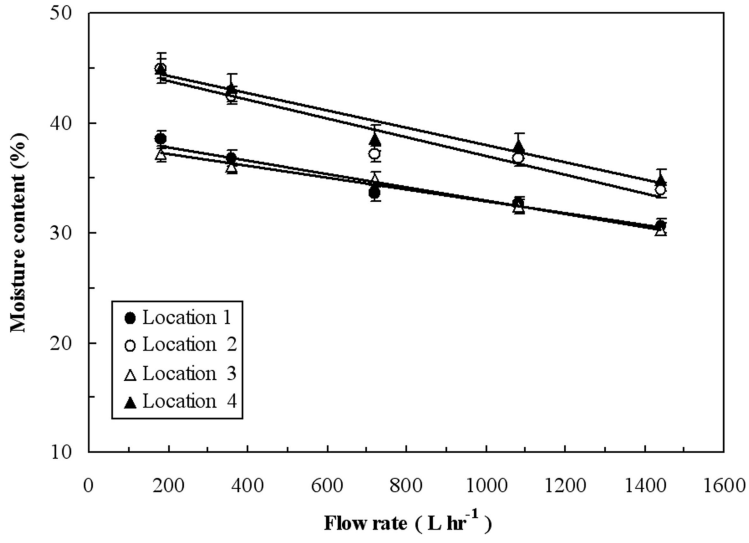

Figure 7. Change of moisture content at the different sampling ports in the two-stage biofilter under various gas flow rates. The liquid recycling rates for both stages were at 5 L min^{-1}. The error bars indicate the standard deviation.

the support material. High moisture generally results in the formation of stagnant zones with diffusion limitations and possible anaerobic conditions or increased pressure drop, and low moisture leads to low microbial activity, making removal of pollutants difficult.[19] Yang and Allen[2] suggest that a moisture content >30% in a biofilter would not materially affect H_2S removal. Below that level, however, the removal efficiency decreases proportionately. Thus, appropriate moisture content in the biofilter system would favor efficient removal of waste gas. The moisture contents of GAC at the different sampling ports in the two-stage biofilter under various gas flow rates and a constant liquid recycling flow rate were measured, and the results are shown in Figure 7. The profiles of the moisture content in the first stage (locations 1 and 2) and in the second stage (locations 3 and 4) showed a significant gradient change in the axial direction of the filter bed. This phenomenon might have resulted from both the force of gravity and the downward direction of gas flow. However, the GAC moisture contents in locations 1 and 3, or in locations 2 and 4, were not different ($P > 0.05$). In other words, similar locations in the first stage and second stage contained similar moisture content. The data indicated that increasing the gas flow rate would result in declining moisture content. The moisture content in the two-stage biofilter was >30% even as the flow rate reached 1440 L hr^{-1}, which may be because of medium inflow being periodically recycled to the system or to the water retention capacity of GAC itself. Additionally, the moisture contents at locations 1 and 3 were periodically determined during the 210-day operation, and results revealed that an average moisture content of 40% was achieved (data not shown). The moisture content was <65% of what a traditional compost biofilter requires to maintain microbial growth in its system.[31] Hence, the two-stage biofilter using GAC as packing material is energy saving.

Pressure Drop

Monitoring of the pressure drop across the biofilter helped us understand the condition of the medium in a long-term operation. In addition, the pressure drop of the biofilter is a key factor in biofilter life. The aging of the compost-based system resulted from acidification or biooxidation, and its disproportionate distribution resulted in a short circuiting of the gases stream accompanied by low removal efficiency.[31] The compression of the medium in the biofilter is because of gas flow and liquid flow, which results in the occurrence of high pressure drop.[32] Because pressure drop is an important factor in the operating cost of the biofilter, the relationship between pressure drop and gas flow rate is crucial to effectively operating the biofilter system.[28] The influence of gas linear velocity on pressure drop is shown in Figure 8. In this experiment, the gas linear velocity was raised gradually from 16 to 127 m hr^{-1} (gas flow rate: 180-1440 L hr^{-1}) after day 160. When the operation reached the steady state (the variation within ±3%) in ~10 days, a new linear velocity was introduced. Inspection of Figure 8 shows that pressure drop of the system increased with increasing gas linear velocity, and both had a good linear

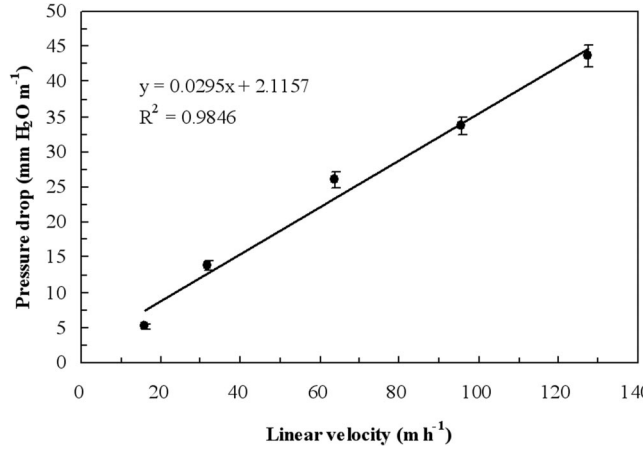

Figure 8. Effect of gas linear velocity on pressure drop across the two-stage biofilter. The data were obtained at 30 ± 2 °C after continuously operating 160 days. The error bars indicate the standard deviation.

relationship (r^2 = 0.9846). The pressure drop range, from 5.12 to 43.65 mm H_2O m filter height^{-1}, below the 250 mm H_2O m^{-1} suggested by biofilter operating guidelines, is very acceptable.[33] When the pressure drop is >250 mm H_2O m^{-1}, the medium must be replaced as previous studies have suggested.[11] In this system, the accumulation of S^o (metabolic product of H_2S biodegradation) could result in the clogging phenomenon. In fact, the relatively low pressure drop compared favorably with the 48–120 mm H_2O m^{-1} of other studies under similar operating conditions.[7,11,14,34,35] The low pressure drop results may be attributed to the small size of the sulfur particles. Thus, the system did not require extra maintenance like chemical rinsing, backwashing, nutrient limitation, or directionally switching operation to avoid clogging for a long time,[36–40] and the two-stage biofilter packed with cell-laden GAC was very economic.

CONCLUSIONS

The results of this study demonstrate that a two-stage GAC biofilter can indeed improve NH_3 removal from waste gases containing high H_2S concentrations. Because the first biofilter inoculated with *T. thioparus* can effectively remove H_2S, the effect of H_2S concentration on nitrification in the second biofilter was avoided. The strategy of using a two-stage biofilter inoculated with sulfur-oxidizing bacteria and ammonia-oxidizing bacteria, respectively, has proven effectual. It provides a new direction to purify gas mixtures in cases in which the biofilter's performance is hindered by the interaction of these gasses. In the first phase (0–70 days), a 98% removal efficiency for NH_3 was achieved when the NH_3 and H_2S gas mixture was <180 ppmv. In the second phase (71–210 days), a 100% removal efficiency for NH_3 was achieved when 60 ppmv of NH_3 and 200–300 ppmv of H_2S gases were introduced. These results bettered those of past research in this area. Additionally, the dominant inoculated bacterial community's ability to exist for 210 days in the single biofilter provides evidence for high NH_3 and H_2S removal efficiency. Therefore, the results of this study suggest that the two-stage biofilter has significant potential to treat NH_3 gas from mixed-waste gases containing high H_2S concentrations.

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